

Cesium-137: Radio-Chemistry, Fate, and Transport, Remediation, and Future Concerns

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In recent years, considerable interest has arisen with regard to the fate and transport of radionuclides such as iodine-131 (¹³¹I), cesium-134 (¹³⁴Cs), and cesium-137 (¹³⁷Cs) in aquatic environments. ¹³⁷Cs is an important indicator of radioactive pollution in aquatic environments. The transport and fate of anthropogenic ¹³⁷Cs is related to the chemical properties of ionic Cs (Cs⁺), which generally dictates a high degree of mobility and bioavailability of this radionuclide. The transport of ¹³⁷Cs and its partitioning between abiotic and biotic components of aquatic ecosystems are complex processes that are considerably affected by a number of factors such as mineralogical composition of suspended and bottom sediments and the characteristic geochemistry of water. These factors influence sorption and desorption kinetics of ¹³⁷Cs and the transport of particulate ¹³⁷Cs. Nevertheless, substantial evidence suggests that direct biological ¹³⁷Cs accumulation from the aquatic environment occurs readily in micro-organisms and aquatic plants. The evidence discussed in this work indicates that ¹³⁷Cs is continuously re-circulated in biological systems for many years following a pulse of contamination. Possible remediation

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methods for the contaminated aquatic systems are also discussed. This review provides guidelines for future work plans for the study of the fate and transport of ^{137}Cs in the aquatic environment in the wake of Fukushima Nuclear Power Plant disaster in 2011 and to provide answers to the urgent questions with respect to strategies for mitigating contamination and reducing radiation exposure for people living in the most affected regions of the world.

KEY WORDS: accumulation, bioconcentration, bottom sediments, desorption, remediation, sorption, suspended solids

1. INTRODUCTION

The distribution of such artificial radionuclide species as ^{131}I , ^{134}Cs , ^{137}Cs , $^{239,240}\text{Pu}$, ^{238}Pu , and ^{241}Am in the current environment depends on the sources (e.g., nuclear weapons tests, accidents at nuclear facilities, submarines and aircraft and leaching from radioactive waste disposal facilities), the release conditions and the subsequent transformation processes (Nuclear Emergency Response Headquarters, 2011; Nuclear Energy Agency, 2002; Valles et al., 2009). In recent years, considerable interest has arisen with regard to the fate and transport of radionuclides in the aquatic environment, especially after the Fukushima Daiichi Nuclear Power Plant (FNPP) disaster on March 11, 2011. This incident led to the release of large amounts of radionuclides into terrestrial, aquatic and marine environments worldwide (Al-Masri et al., 2003; HELCOM, 2003; Szefer, 2002; United Nations Environment Program, 1992). For example, early estimates suggest that $\sim 150 \times 10^{15}$ Bq of ^{131}I , $\sim 100 \times 10^{15}$ Bq of ^{134}Cs , and $\sim 13\text{--}50 \times 10^{15}$ Bq of ^{137}Cs were released into the atmosphere within a month after the incident (HELCOM, 2009; Ikäheimonen et al., 2009; Nielsen et al., 1999; Zalewska et al., 2006). Because these anthropogenic radionuclides have migrated and diffused through the atmospheric and aquatic environments, a nationwide survey was conducted to monitor their dispersion, understand the resulting contamination levels and mitigate the human health and environmental risks. Scientific knowledge and effective remediation methods for radionuclides are thus necessary to maintain and recover the safety of our society, environment and ecosystems.

Among these artificial radionuclides, the long-lived ^{137}Cs is an important indicator of radioactive pollution in aquatic environments. This importance results from its large mass production during nuclear fission events and longer half-life (30.07 years) compared with other radiocesium isotopes, such as ^{134}Cs (half-life 2.06 years; Littler et al., 1980; Lokobauer et al., 1998; Nuclear and Industrial Safety Agency, 2011; Wernsperger et al., 2004). $^{137}\text{Cesium}$ in the environment originates from a variety of sources. The largest single source was fallout from atmospheric nuclear weapons tests in the 1950s and

1960s, which had dispersed and deposited ^{137}Cs worldwide. However, much of the ^{137}Cs from nuclear weapons testing has now decayed (Great East Japan Earthquake Taskforce, Science Council of Japan, 2011; International Atomic Energy Association, 1996). Nuclear reactor waste and accidental releases, such as those from the FNPP disaster and the Chernobyl accident in the Ukraine, are the major sources of ^{137}Cs in the current environment. Spent nuclear fuel re-processing wastes may introduce small amounts of contamination into the environment (Directorate of Radiological Protection and Human Health, 2011). Europe is actively reprocessing spent nuclear fuel (Al-Masri et al., 2003; Giani et al., 1997; Szefer, 2002; United Nations Environment Program, 1992). However, the United States does not reprocess spent nuclear fuel at this time (Franić et al., 1993; Turekian et al., 1977). Although hospitals and research laboratories do generate waste containing ^{137}Cs , these materials normally do not enter the environment (Pieńkowski et al., 1987). Occasionally, industrial instruments containing ^{137}Cs are lost or stolen (Leon et al., 2011), and anyone who unwittingly handles these materials may be exposed. Because these devices typically consist of metal, they may be considered as scrap and sold for recycling. Additionally, if these materials find their way into a steel mill and are melted, they can cause significant environmental contamination. The materials may be also discarded and sent to a municipal landfill or sold for other reasons. Therefore, these devices should be considered potentially hazardous (Lozano et al., 2011). When atmospheric fallout is deposited onto land areas, the radioisotopes attach to fine-grained sediments transport into marine environments (Długosz et al., 2010; Długosz-Lisiecka et al., 2012; Kapała et al., 2002). In coastal areas, the majority of ^{137}Cs originates from atmospheric deposition, and the distribution of ^{137}Cs in the marine environment is related to the temporal pattern of fallout of ^{137}Cs onto the earth's surface on a global scale (Currie, 1999; Dale et al., 2001; Grabowski et al., 2010; Manolopoulou et al., 2011).

In addition to its relative abundance, ^{137}Cs possesses characteristics that enhance its importance as a contributor to radiation doses. For example, this material has a half-life (~ 30 years), emits relatively high-energy beta particles and its rather short-lived daughter, $^{137\text{m}}\text{Ba}$, emits strong gamma rays. Due to its chemical properties, ^{137}Cs is readily transported through the environment and food chains (Chino et al., 2011; HELCOM-MORS, 2010; Turekian et al., 1977). With respect to its biogeochemical properties, ^{137}Cs is readily soluble in water, and, in solution, it can be efficiently taken up by plants and assimilated by animals because of its chemical similarity to the essential nutrient potassium (Popović, 1978, RC Hoetzlein, 2011; Sobotovitch, 2001; Yasunari et al., 2011). The primary factor limiting the transport of ^{137}Cs to humans and other living organisms is its notably strong tendency to attach (sometimes irreversibly) to common clay minerals found in most soils and sediments (Bauman et al., 1992; Marović et al., 2010). On the other hand, being a monovalent cation it would also tend to sorb weakly to most mineral

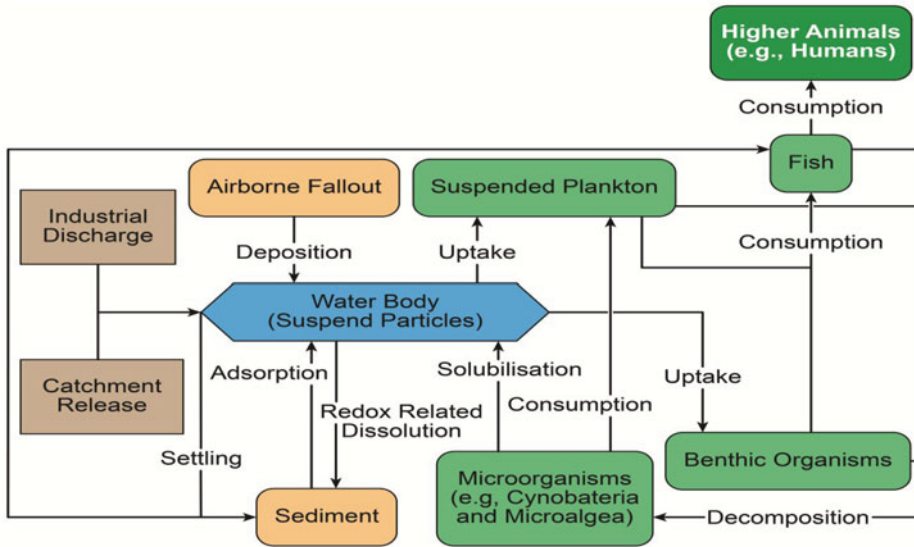


FIGURE 1. ^{137}Cs recycling in aquatic ecosystems.

surfaces, kaolinite, Fe-oxides, 2:1 clays, but very strongly to illites or hydroxyl-interlayered vermiculite (Franić et al., 2006; Tang et al., 2000). Over time, ^{137}Cs increasingly binds with soil and sediment particles, which reduces its transport to plants and animals, but the gamma-ray emissions of its daughter $^{137\text{m}}\text{Ba}$ may still produce significant external radiation exposure (Marović et al., 2010). Furthermore, small soil particles containing ^{137}Cs can move through the environment via wind and water erosion, and re-suspended airborne materials with sufficiently small particle diameters can be inhaled by humans and other animals (Franić et al., 2006; Tang et al., 2000). Figure 1 shows a schematic diagram summarizing the major components and processes of ^{137}Cs transport and transformation in aquatic ecosystems.

The geographic distribution of ^{137}Cs is influenced by numerous factors. These factors include the locations and magnitudes of release to the environment, the medium (air, water, soil) into which the material is released, the flow patterns of air or water and the deposition processes occurring from those media (Franić et al., 1993; Porcelli et al., 2001; Tang et al., 2000). The global distribution of ^{137}Cs on land is influenced mainly by its release to the atmosphere and mechanisms that cause deposition from air to surfaces, such as soil, vegetation, water, rock, ice and man-made structures (Abraham et al., 2000; Liu et al., 2000). At a given latitude, the deposition of particulate aerosols increases in proportion to the concentrations in the column of air through which precipitation falls as well as to the amount of rainfall (Franić et al., 1993). For example, the areas most impacted by fallout from the Fukushima accident were located either quite close into the reactor or

in places where rainfall occurred when the plume of airborne radioactivity drifted over the geographic area (Kumblad et al., 2006). Other factors can also play a part and are described in the body of this article. In the case of release to streams, the ultimate fate of the ^{137}Cs contamination depends on the characteristics of the stream, including the velocity, dilution and sediment load, among others (Carlson, 1990; Lepicard et al., 2004; Takemura et al., 2011). The ^{137}Cs in solution or attached to small particles can travel hundreds of kilometers if the stream flows are sufficient (Visible Information Center, 2011). Water drawn from streams for irrigation can result in the transfer of contaminated material from the stream to the land. Conversely, runoff can carry small soil particles containing ^{137}Cs from the land to waterways (Deutscher Wetterdienst, 2011; Kaplan et al., 2001).

Because aquatic contamination with ^{137}Cs is a major concern in many regions of the world (e.g., New York, Alaska, Hawaii, Oregon, California, Montreal, Austria; Jarvis et al., 2001; Lindley et al., 1996; Rushton, 2003; Sexton et al., 2000; Waller et al., 1999) updated knowledge of the fate and transport of ^{137}Cs is important for devising effective strategies and developing techniques for agricultural countermeasures and phytoremediation to minimize the transfer of ^{137}Cs from the aquatic ecosystem to humans. Five years have passed since the National Council for Radiation Protection and Measurements (NCRP; 2007) published a report dealing specifically with the behavior of ^{137}Cs in the environment, and during that period a large body of research has been published. After the Fukushima disaster, there was an urgent need to study the behavior of ^{137}Cs in the ecosystem. Thus, a review that collects a reasonable fraction of the rather extensive literature and summarizes the current knowledge of how this radionuclide behaves in the aquatic ecosystem can be highly useful for future risk assessment and for management decisions concerning radioactively contaminated areas, regardless of the source of contamination (Unterweger et al., 1992). Much of the earlier work on ^{137}Cs in the environment was motivated by scientific curiosity and by general concerns over the health and environmental impacts of global fallout from nuclear testing, and later concerns emphasized the safety of nuclear reactors used to produce electricity. However, in the 1970s and early 1980s, research funding for radioecology (a field that deals with studies of how radioactive substances interact with nature; how different mechanisms affect the substances' migration and uptake in food chain and ecosystems began to decline, particularly in the United States. The Chernobyl accident in 1986 and the recent FNPP disaster have spawned many new studies in radioecology, primarily in Japan, the United States, the former Soviet Union, and other European countries. Because of its abundance and ease of measurement, ^{137}Cs has most likely received a good amount of study than all of the other radionuclides combined. This work adds to the significant body of literature from earlier studies. As a result of these changing drivers of environmental research on ^{137}Cs , there has been a general shift in focus from the

basic science aspects prior to the mid-1980s to more applied aspects since that time.

In brief, this article reviews the current understanding of the fate and transport of ^{137}Cs in aquatic ecosystems to gather information for effective management practices and future research on the fate and transport of ^{137}Cs in aquatic ecosystems. Similarly, information on the distribution and fate of ^{137}Cs can also contribute to an understanding of the transport processes of other radioactive contaminants that behave similarly and to provide answers to urgent questions with respect to strategies for mitigating contamination and reducing radiation exposure for people living in the most affected regions.

2. RADIO-CHEMISTRY OF ^{137}CS

Cesium is an alkali metal and is a chemical analogue to potassium (Filipović-Vinceković et al., 1991). $^{133}\text{Cesium}$ is the only stable isotope of cesium, although this element can exist in over 20 isotopic forms, including the complete series from ^{123}Cs to ^{144}Cs , with the exception of ^{124}Cs (United Nations Scientific Committee on the Effects of Atomic Radiation, 1982). The nuclear evolution has resulted in the large-scale release of Cs radioisotopes into the environment, in particular, ^{134}Cs (half-life ~ 2.07 years) and the long-lived radionuclide ^{137}Cs (half-life ~ 30.2 years; International Atomic Energy Association, 1986; United Nations Scientific Committee on the Effects of Atomic Radiation, 1988). ^{134}Cs is produced by neutron activation of the stable end product of the 133 atomic-weight fission chain (Franić et al., 2007). ^{137}Cs is a fission product that has short-lived precursors such as ^{134}Cs . These precursors are significant contaminants because of their high fission yield and their relatively long half-lives (Poet et al., 1972). The fission yield of ^{137}Cs in nuclear reactions is relatively high; approximately six atoms of ^{137}Cs are produced per 100 fission events (Tokuyama et al., 1998). ^{137}Cs decays by beta decay either to stable ^{137}Ba or to a meta-stable form of barium ($^{137\text{m}}\text{Ba}$; Jian et al., 2012). The meta-stable isotope ($^{137\text{m}}\text{Ba}$) is rapidly converted to stable ^{137}Ba (half-life of approximately 2 min) accompanied by gamma-ray emission with an energy of 0.662 MeV, as shown in Figure 2. The first beta decay mode that forms $^{137\text{m}}\text{Ba}$ accounts for roughly 95% of the total intensity, and the second mode accounts for approximately 5%. Radioactive ^{134}Cs primarily decays to stable ^{134}Ba by beta decay accompanied by gamma ray emission or less frequent decay to stable ^{134}Xe by electron capture (EC) accompanied by a single gamma ray (Aoyama et al., 2008; Polish National Atomic Energy, 2011). ^{137}Cs is a fission product of both U- and Pu-reactors. The major signal used to identify ^{137}Cs is gamma radiation at 661 keV, which is the product of beta decay to $^{137\text{m}}\text{Barium}$ ($^{137\text{m}}\text{Ba}$; Brenner et al., 2003; Jia et al., 2000).

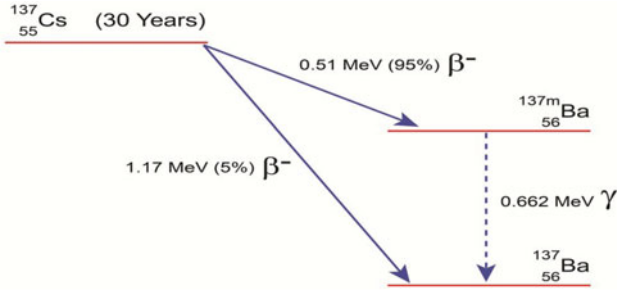


FIGURE 2. The decay scheme of ^{137}Cs .

3. DISTRIBUTION OF ^{137}CS IN AQUATIC ECOSYSTEMS

The release of ^{137}Cs into the aquatic ecosystem generally occurs through association with suspended soil particles of different sizes and mineralogical composition that enter aquatic ecosystems, which considerably affects their transport and bioavailability (Endo et al., 2012; Qin et al., 2012). $^{137}\text{Cesium}$ contaminates bodies of water not only directly via deposition from the air and discharge as effluent but also indirectly via washout from catchment basins (Hirose et al., 2008). Because the ^{137}Cs that contaminates large bodies of water is quickly redistributed, it tends to accumulate in bottom sediments, benthos, aquatic plants and fish (International Atomic Energy Association, 2001), and the main pathways for potential human exposure may occur directly through the contamination of drinking water or indirectly from the use of water for irrigation and the consumption of contaminated fish or plants (Japanese Ministry of Education Culture Sports Science and Technology, 2011a). Because the contaminating radionuclides tend to dissipate from water rather quickly, the scenarios described previously represent only the initial fallout phase. In the later phase, when the contamination washed out from the catchment area reaches the drinking water supplies, human exposure is more likely (Japan Ministry of Land Infrastructure Transport and Tourism, 2011). Table 1 provides a summary of the major contributors to ^{137}Cs in the aquatic environment.

Radioactive contamination of ^{137}Cs in the aquatic system is caused by three main factors: global fallout, discharge from reprocessing plants and fallout after incidents such as the Chernobyl accident in April 1986 and the Fukushima nuclear incident of 2011 (Bossew et al., 2007; International Atomic Energy Association, 2000). At present, the average activity concentration of ^{137}Cs in the surface water of the Pacific region is estimated at approximately 60 Bq/m^3 , whereas the worldwide average concentration due to global fallout is approximately 2 Bq/m^3 (International Atomic Energy Association, 2001; International Commission on Radiological Protection, 2003). The infamous Chernobyl nuclear power plant accident in Ukraine on

TABLE 1. Sources of ^{137}Cs in the aquatic environment

Type of release	Sources	Cumulative amount of ^{137}Cs released ($\times 10^{15}$ Bq)			
		World	Reference	Asia-Pacific	Reference
Weapon tests	a	1300	(Eisenbud, 1987)	500	(Koulikov et al., 1992)
	b	650–1100	(Christopher, 1987)	580	(Lloyd et al., 1973)
	c	900–1150	(Dominik and Span, 1992)	595	(Lloyd et al., 1973)
Accidental	Sellafield	85	(Dominik and Span, 1992)	26	(Lloyd et al., 1973)
	Windscale	82	(Čonkić et al., 1990)	20	(Dominik and Span, 1992)
	Chernobyl	70	(Cambray et al., 1987)	25	(Dominik and Span, 1992)
	Fukushima	88	(Chossudovsky, 2012)	78	(Chossudovsky, 2012)

April 26, 1986 (the second most significant large-scale fallout source of environmental radioactive contamination in the Arctic), is another significant source of large-scale global fallout of radioactive cesium. This accident was a consequence of uncontrolled fission in the reactor followed by a powerful explosion and fire (Brechignac et al., 2003; Jasiulionis et al., 2006). The radioactive materials released were carried away by air currents in the form of gases and dust particles (Jasiulionis et al., 2007). As a result, a total of 85 PBq of ^{137}Cs was added to the atmosphere (Medici, 2001; Japanese Ministry of Education Culture Sports Science and Technology, 2011b).

The earthquake and the subsequent tsunami on March 2011 resulted in the release of radionuclides from three boiling water reactor (BWR) units of the Fukushima Daiichi power station in Japan, which are considered to be the major contributors of artificial radionuclides to the aquatic ecosystem. The main source was the direct discharge of contaminated water from the plant, which lasted until approximately April 8. A lesser discharge that impacted the ocean involved radionuclides discharged into the atmosphere between March 12 and 22 (Japanese Ministry of Education Culture Sports Science and Technology, 2011c; Lee et al., 2008; Morino et al., 2011; Real et al., 2002). In the immediate vicinity of the plant, the concentrations in seawater reached several tens of thousands of Bq/L for ^{134}Cs and ^{137}Cs and even exceeded 100 000 Bq/L for ^{131}I near the end of March and early April, as shown in Figure 3 and Abraham et al. (2000) and Chino et al. (2011). The radioactive release at sea was the largest one-time contribution of artificial radionuclides to the marine environment ever observed (Stohl et al., 2011). There was a strong dispersion of radionuclides from the site of Fukushima (from Russia, to Los Angeles) due to its geographic location in the Pacific Ocean near one of the strongest water currents in the world (Japanese Ministry of Education Culture

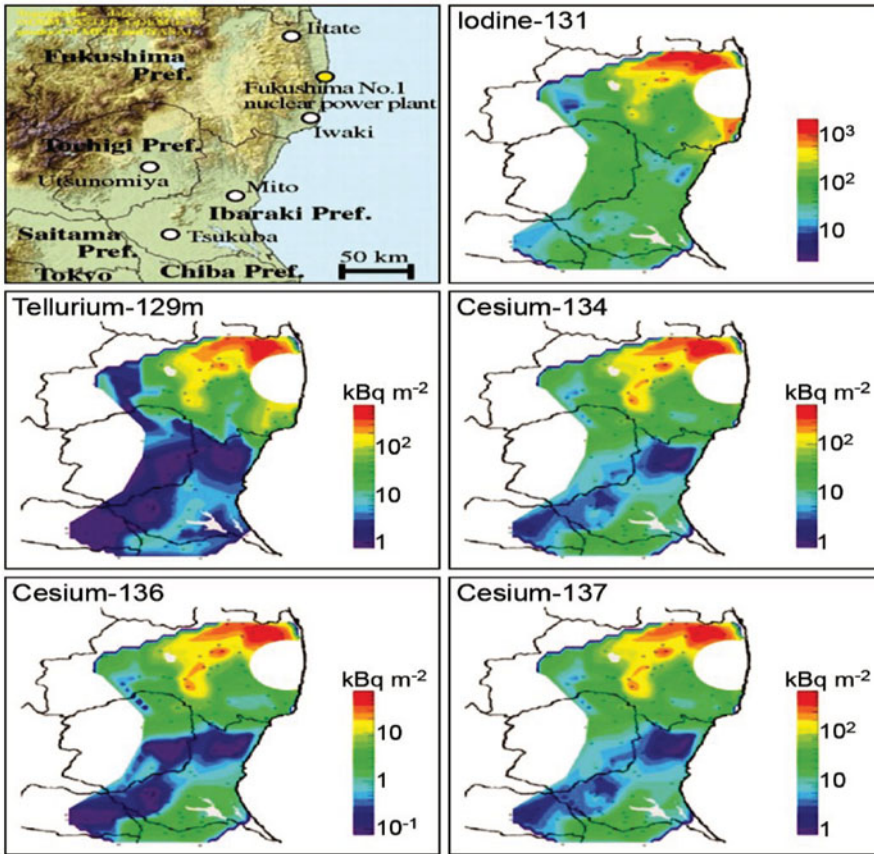


FIGURE 3. Distribution of radionuclide deposition (^{131}I , ^{129}Te , ^{134}Cs , ^{136}Cs , and ^{137}Cs) from the Fukushima Daiichi accident (Kinoshita et al., 2011).

Sports Science and Technology, 2011d). The measurement results obtained from the seawater and coastal sediments suggest that the consequences of the accident, in terms of radiation protection, produced weak pelagic species beginning in the fall of 2011 (low concentrations in seawater and limited sediment storage). Similarly, the surface soil near the FNPP is now heavily contaminated with depositions of ^{137}Cs at levels of more than 100,000 MBq km^{-2} (United Nations Scientific Committee on the Effects of Atomic Radiation, 2000) compared with the ^{137}Cs level of $\sim 270 \text{ Bq km}^{-2}$ before the incident (Jasiulionis et al., 2006).

4. TRANSPORT OF ^{137}CS IN AQUATIC ECOSYSTEMS

The transport of radionuclides through aquatic systems is partially dependent on the physical and chemical properties of the contaminant and on

the rock and sediment characteristics (Taira et al., 2011). ^{137}Cs strongly interacts with particulate matter suspended in water, and the bottom sediments and suspended particles show comparatively lower levels of mobility (Marcuilioniene, 2002). The experimental evidence also demonstrates that the horizontal migration of the radionuclide through surface waters is of paramount importance. Horizontal radionuclide fluxes in surface water are generally high and often determine the spatial extent of the contamination (Nedveckaite, 2004). In general, the vertical migration of radionuclides to the underlying groundwater aquifers is comparatively weak. In fact, soils were found to act as effective filters for radionuclide transport to underground water (Sanzharova, 2009). For example, studies carried out in Belarus and by the Russian Federation show that the radiocesium and radiostrontium of Chernobyl fallout were still retained by the upper layers of the soil humus horizon 14 years after the accident (International Atomic Energy Association, 2009).

The vertical migration characteristics of water column were found to depend primarily on the contaminant levels and their forms (particles or aerosols), the radionuclide in question, the soil and rock characteristics and the hydrological properties of the water column (Shaw, 2007). It should be also noted that the vegetation cover and soil microbiota might have a strong seasonal and long-term effect on ^{137}Cs mobility and vertical migration through the water column. Naturally occurring radionuclides can also provide valuable insight into the pollutant migration mechanisms in aquatic environments (Japanese Ministry of Education Culture Sports Science and Technology, 2012). A study of the distribution of ^{137}Cs in the water column of a coastal lagoon in Brazil has shown that factors such as the hydrological regime (salinity, pH, etc.) can greatly influence the migration processes. ^{137}Cs is a conservative radionuclide, which means that it behaves as a component of its associated water body (Japanese Ministry of Education Culture Sports Science and Technology, 2011). The initial relative distribution of released ^{137}Cs between the terrestrial and aquatic ecosystems depends primarily on the source of the radioisotope. Additionally, cesium has a relatively low K_d : $\left(\frac{\text{Activity per unit mass solid}}{\text{Activity per unit volume liquid}}\right)$ Bq/L of approximately 3×10^3 ml/g for sediments/waters (Kinoshita et al., 2011).

The interaction and subsequent association of ^{137}Cs with the solid phase can take place via a number of mechanisms (Taira et al., 2011), including the direct uptake of ^{137}Cs at the sediment-water interface, sedimentation with organic matter either after assimilation or through adsorption onto cells, adsorption onto inorganic compounds (e.g., clays and carbonates) or scavenging from solution by iron-manganese oxy-hydroxides, sedimentation with humic matter and direct uptake through assimilation by periphyton or other biota residing on surface sediments. ^{137}Cs is often adsorbed onto specific sites located within the wedge zones of illitic crystallites (Japanese Ministry of Education Culture Sports Science and Technology, 2011; Møller et al.,

2012; Møller et al., 2013). ^{137}Cs fixation (i.e., interlayer collapse of the clay lattice) can subsequently take place, leading to the virtually irreversible fixing of the cesium ion within the clay mineral matrix (Tsukada et al., 2003). Investigations into the geochemical association of ^{137}Cs with soils and sediments have shown a strong association of cesium with the irreversibly bound fraction, which corresponds to an association of ^{137}Cs with primary and secondary minerals, including illite (Sinclair et al., 2011). ^{137}Cs associated with this geochemical phase will not be readily available for biological uptake or re-mobilization. However, the ^{137}Cs molecules associated with the exchangeable, geochemical phases in marine sediment may be available for uptake by biota and prone to desorption processes (Bowyer et al., 2011). In the Irish Sea, for example, studies confirm that significant inventories of ^{137}Cs have remobilized from sediment deposits (Brandt et al., 2002; Central Institute for Meteorology and Geodynamics, 2011).

4.1. Fate of ^{137}Cs in Aquatic Ecosystems

Despite the large proportion of the earth's surface occupied by water and the high mobility and availability of ^{137}Cs in solution, the fate of ^{137}Cs in aquatic ecosystems has been less extensively examined than in terrestrial ecosystems, which may be partly due to the greater contribution of food originating from terrestrial sources to the human diet (Chino et al., 1993; Chino et al., 2011).

The fate of radiocesium in the environment is thus dependent on a highly complex series of interactions that are ultimately governed by biological activity, the prevailing environmental and physicochemical conditions and the chemical behavior of Cs^+ . The relative distribution of Cs^+ between the abiotic and biotic components of the environment is highly variable and dynamic, and the behavior of Cs^+ differs considerably in terrestrial, freshwater and marine ecosystems (Nelson et al., 1971). It is clear that Cs undergoes intensive recycling in the environment, and the relatively long half-life of ^{137}Cs (~ 30 years) indicates that radiocesium can remain a serious problem for many years following a pulse of contamination. ^{137}Cs activities are lower in sediments than in plant material (Zheng et al., 2012). The concentrations in the various aquatic plants may reflect their morphology because those plants with larger surface areas also possess a greater internal content (Cornell, 1993).

Considerable evidence suggests that organisms in aquatic habitats are more susceptible to ^{137}Cs contamination than their terrestrial counterparts (Davoine et al., 2007). For example, it has been estimated that a 570- to 1500-fold greater accumulation of ^{137}Cs occurs when plants are grown in water as opposed to soil; the variability in this estimate was attributed to differences in the moisture content of the soils (Antonio et al., 1997; Devell et al., 1995; Galmarini et al., 2011). In addition, Polar et al. (1991) reported

TABLE 2. Biological and environmental half-life of ^{137}Cs

Organism	Half-life	Reference
Moss	4.5 years	(Daróczy et al., 1994)
Lichen	5–8 years	(Ellis and Smith, 1987)
Grass	14 days	(Cambray et al., 1987)
Plant surface	14 days	(Komarov et al., 1983)
Hen	1.5 days	(Ekman, 1961)
Cow	3 days	(Cragle, 1961)
Fish	70–300 days	(Koulikov et al., 1992)
Child	57 days	(Lloyd et al., 1973)
Woman	84 days	(Lloyd et al., 1973)
Man	105 days	(Lloyd et al., 1973)
Monomictic lake ^a	1.2 years	(Dominik and Span, 1992)
Meromictic lake ^b	6.7 years	(Dominik and Span, 1992)
Flowing river	1.4 years	(Čonkić et al., 1990)
Airborne dust	270 days	(Wang et al., 2011)

^aMix from top to bottom during one mixing period each year.

^bHas layers of water that do not intermix.

the elevated levels of ^{137}Cs uptake in aquatic plants compared with terrestrial plants occurred when the same Cs-contaminated medium was used as a sediment material in the first instance and as a soil in the second (Haritonidis et al., 1995; Manolopoulou et al., 2011; Sokolov, 2001).

4.2 Bioavailability of ^{137}Cs in Aquatic Ecosystems

The availability of ^{137}Cs for biological uptake is of vital importance for transfer to humans through food webs. To develop proper countermeasures, the short- and long-term bioavailability of ^{137}Cs should be known and taken into account. Particular concern has arisen over the distribution of ^{137}Cs between the abiotic and biotic components of aquatic ecosystems (Norman et al., 2011). The tendency of Cs radioisotopes to accumulate in biological food chains has been further highlighted by several reports suggesting that radioactive Cs is accumulated in preference to stable Cs in certain organisms (Franić, 2005; Giani et al., 1997), and more importantly, in preference to the chemically similar and biologically essential alkali metal potassium (Bowyer et al., 2011). Consequently, ^{137}Cs may eventually accumulate in higher organisms, including humans, in which both lethal and sub-lethal ionization effects may be exerted at the molecular level (Sinclair et al., 2011). It should be noted that the time-scale of ^{137}Cs retention in higher organisms (biological half-lives of generally less than one year; Table 2) dictates that ^{137}Cs accumulation in these organisms does not continue indefinitely, and thus a steady-state internal Cs level will eventually occur during exposure to constant external radiocesium levels (Lozano et al., 2011; Pittauerová et al., 2011). The bioavailability also increases with water depth because the fixation of ^{137}Cs to the soil matrix has not yet been completed at greater depths.

4.3 Distribution of ^{137}Cs in the Abiotic Components of Aquatic Ecosystems

4.2.1 DEPOSITION

^{137}Cs first enters the surface of the aquatic system as direct deposition and later as runoff from the surrounding drainage areas (Bolsunovsky et al., 2011). The runoff of fallout nuclides from the land to the aquatic system depends on many factors associated with the type and properties of the soil in the drainage area (Morino et al., 2011). In the course of time, particle-bound cesium is carried by erosion and runoff from the drainage area to the aquatic system (Stohl et al., 2005). The high activity concentrations of ^{137}Cs in the water phase and the relatively low concentrations in the bottom sediments of the lakes indicate that the runoff process of cesium from the drainage areas to the aquatic system has become relatively slower due to the particle-bound cesium in the drainage areas (Aoyama et al., 2011). The oligotrophic nature of these aquatic systems is surely one reason why cesium remains longer in the water phase.

Natural sediments usually act as a sink for various contaminants, but they may also become a source under the drastically changing environmental conditions in the fresh and saline water mixing zones (Buesseler et al., 2011). The solubility, potential mobility and bioavailability are responsible for the harmful effects of cesium on animals and the environment. In addition, a variety of factors (i.e., water pH, ionic strength, competitive sorption, and complexation with inorganic and organic ligands) can influence the sorption process and thereby the mobility of the radionuclides (Hamajima et al., 2004). However, it is now generally recognized that the sorption of ^{137}Cs by soil and sediments is mainly determined by specific sorption onto illite clay minerals, and such parameters as pH and organic matter were found to play less important roles in the adsorption of Cs (Honda, 2012). Three types of ^{137}Cs binding sites for sediment clay components have been identified experimentally: (a) sorption to surface and planar sites (Cs^+ is generally exchangeable), (b) sorption to wedge sites (Cs^+ exchange limited only to cations of similar size and charge), and (c) sorption to interlayer sites (Cs^+ is not readily exchangeable). The selective adsorption of Cs by clay minerals has been attributed to the large ionic radius, uncomplexing nature and low hydration energy (Inoue et al., 2012; Mori et al., 2011). Although cations with similar charges and ionic radii are expected to compete with cesium, the sequence of the sorption ability of the alkali elements of $\text{Na}^+ < \text{K}^+ < \text{Rb}^+ < \text{Cs}^+$ is in good agreement with the sequence of the effective ionic radii of the alkali elements and the sequence of the single ion hydration enthalpies of the alkali elements (Japan Society of Civil Engineers, 2011).

In near-shore waters, the sediments may act as a Cs^+ sink (Tsumune et al., 2012). Furthermore, the deposition of ^{137}Cs onto aquatic sediments may be facilitated by the settling of Cs-associated solids in the water column,

particularly because these materials appear to have a higher affinity for Cs^+ than for K^+ in many cases (Hinton et al., 2002). Fowler et al. (1987) concluded that a major route of the rapid downward migration of radioactivity in seawater following the Chernobyl accident occurred via dense fecal pellets from zooplankton that had previously grazed on contaminated phytoplankton and other smaller particulates (International Atomic Energy Association, 2004). ^{137}Cs preferentially binds to fine-grained particles and becomes deposited in sediments associated with high rates of settling of these fractions (Bellenger et al., 2008; Fisher et al., 1999). Thus, ^{137}Cs can be used as a tracer for sediment dating (Bostick et al., 2002). For instance, Mihai (2003) correlated peaks of radioactivity in sediment samples from the river Danube with ^{137}Cs from nuclear weapons tests in the 1960s and from Chernobyl in 1986. However, it should be emphasized that such applications are only valid if the radiocesium is deposited rapidly from the water column to the sediment and is not subsequently subjected to significant remobilization or redistribution (Cha et al., 2006). Thus, as a result of the pre-depositional mixing of discharges from the Sellafield site in Cumbria, England (where a lag of several years occurred between the radionuclide release and maximum sediment accumulation), the sediment profiles provide a record of the time-integrated Sellafield discharge rather than of the annual fluctuation (Cremers et al., 1988). These findings can be further complicated by the redistribution of contaminated silt and radionuclide dissolution (Fan et al., 2012). The former process may result in the transport of particle-associated radiocesium to intertidal areas, thus increasing the likelihood of human contamination (Giannakopoulou et al., 2007).

4.2.2 MOBILITY AND DISTRIBUTION

The harmful effects of cesium on animals and the environment are evident considering its properties of unlimited solubility, potential mobility and high bioavailability. The mobility of the radionuclide in the aquatic environment is governed by complex physicochemical interactions, which depend largely on the properties and characteristics of the nuclide, the water body and the surface area of the charged particle (Giannakopoulou et al., 2007). The mobility and fate of radiocesium in the aquatic environment depend to a great extent on its speciation and transformation. The removal processes for a portion of the water-soluble radiocesium released to the environment are usually related to their sorption onto soil and sediment particles (Honda et al., 2012). Most of the ^{137}Cs that is deposited in the open water remains in solution indefinitely (Tanaka et al., 2012) in cationic form (Kato et al., 2012). For example, one month after the main pulse of fallout from Chernobyl entered the sea around Monaco in 1986, only 0.2% of the ^{137}Cs in the water body had exceeded a depth of 200 m (Kinoshita, 2011). In the open sea, ^{137}Cs is considered to be distributed primarily by water circulation processes, and consequently, ^{137}Cs may be used as a tracer of water movement

(Nakamaru et al., 2007; Ohno et al., 2012; Tanaka et al., 2012). However, the distribution of ^{137}Cs between the soluble and sediment-bound phases is variable and depends on the vertical stability and depth of mixing of the sea, processes that allow the radionuclide to come into contact with sediments (Tsukada et al., 2002). In surface waters, radiocesium is present in different physicochemical forms (water soluble, exchangeable, bound to Fe-Mn, persistently bound, and residual) varying in molecular size, charge, and density. The importance of the complexation of long-lived radionuclides with naturally present organic materials in the aquatic environment is well recognized. The distribution of ^{137}Cs in natural waters also depends on the water chemistry (pH, ionic strength, inorganic components, and organic properties) and transportation processes (Watanabe, 2012; Yasunari et al., 2011).

^{137}Cs is relatively immobile in lake sediments, although a certain amount of dispersion may occur via the physical mixing of sediments (by biota and water currents) or diffusion in the sediment pore waters. Sediments of high clay content can effectively adsorb cesium, and this process largely determines its immobilization in sediment (Yoshida, 2012; Zachara et al., 2002). The total amount of ^{137}Cs in the sediment depends on the type and properties of the soil of the drainage area as well as biological factors and processes such as particle production, sedimentation rate, re-suspension and the removal of particles from the water column (without flowing water masses). Approximately 95% of the ^{137}Cs introduced into the aquatic environment may be rapidly adsorbed and retained in the sediment, but this sorption can be reversible in certain sediments (Stohl et al., 2012). The presence of other monovalent cations can also markedly influence the distribution of ^{137}Cs in aquatic systems. The ions NH^+ , H^+ , K^+ , and Na^+ all have the potential to displace sediment-bound Cs^+ (Yasunari et al., 2011). Indeed, Davison et al. (1993) concluded that the inundation of coastal freshwater basins by seawater (high Na^+ concentration) represented an important route by which sediment-bound Cs could be rapidly reintroduced into the water column, although the mobilization was more likely to occur from sinking particles than settled sediment. In addition, the slow leaching of ^{137}Cs from contaminated catchments can maintain high ^{137}Cs levels in freshwater lakes over several years (Campbell, 1986). Kashparov et al. (2003) demonstrated that the extended retention of ^{137}Cs in the water column of a Cumbrian (England) lake after the Chernobyl accident was most likely due to continued release of ^{137}Cs from catchment soils, particularly those composed of fibrous peat (Yasunari et al., 2011). Catchments (and sediments) rich in illitic clay minerals, to which Cs^+ binds almost irreversibly, will retain Cs more strongly, resulting in lower Cs^+ concentrations in the water column.

The mobility and distribution of ^{137}Cs in sediments are governed by essentially the same processes as those occurring in soils. ^{137}Cs binds preferentially to clay rather than sandy sediments in marine ecosystems (Wong, 2000). The sediment accumulates the radionuclide as a result of scavenging

and settling processes in the water column. Adsorption of Cs^+ to sediments actually appears to be biphasic. Skei et al. (1979) observed mean Cs^+ sorption times to aquatic system sediments of 2.4 and 42 days and suggested that these values may have been related to distinct modes of Cs^+ trapping in sediments (e.g., in the interlayer regions of vermiculite and at the frayed edges of illites and micas; Jacobs, 2000; Nikolova et al., 2000). Cesium-137 is generally considered to bind most readily to Al-rich particles (Chen, 2006) and particularly the illites (Ulsh et al., 1999) of sediments. The association of Cs^+ with sediments results in a massive concentration of the radionuclide compared with the lower concentrations prevalent in the surrounding waters (Forsberg et al., 2000). However, binding within sediments is not necessarily irreversible. For example, Cs is weakly bound and relatively mobile in organic-rich lake sediments (Yang, 2000). Furthermore, Alberts et al. (1979) observed a seasonal cycling of Cs^+ between water and sediment in a monomictic lake system, which coincided with periods of thermal stratification and cyclical precipitation and dissolution of ferric oxides (Nikolova et al., 2000). Similar interactions have been reported by Dominik and Span (1992), who related periods of intensive ^{137}Cs recycling in Lake Lugano in Switzerland to the prevailing redox conditions (Seaman et al., 2001). These authors attributed the slow overall half-removal times of ^{137}Cs to sediment (6.7 years in meromictic lake; 1.2 years in monomictic lake; Table 2) to these redox-related processes. In addition, Shestopalov (2002) provided evidence that the observed Cs releases from freshwater lake sediments were due to periods of hypolimnetic anoxia during the summer months (Camps et al., 2003). In certain cases, seasonal variations in freshwater ^{137}Cs activities may be partly attributable to climatic changes resulting in a release of ^{137}Cs from melting snow or ice in the surrounding catchments (Xiong et al., 2005).

Therefore, the concentration of ^{137}Cs in marine sediment may be also influenced by particle size, the mineral composition and the content of organic matter in marine sediments and the relationship between organic matter and ^{137}Cs in the sediments (Henley et al., 2000; Yasunari et al., 2011). In the sea, ^{137}Cs can deposit on the surface of marine sediments via a variety of mechanisms, including fixation on suspended matter and sedimentation, direct precipitation of colloidal forms, direct fixation by adsorption and deposition of organic waste (Bunzl, 2002). The content of organic matter in coastal sediments is generally higher than that of pelagic sediment. The organic fraction present is important in terms of the binding and fixation of ^{137}Cs to sediments. Organic matter is also important in sea-bed-water-column transfer processes due to the number of functional groups found on organic molecules (Butkus et al., 2009). ^{137}Cs is also strongly adsorbed to clay particles, which have large surface areas and fine particle sizes, and the adsorbed ^{137}Cs is virtually nonexchangeable in the sequential extraction procedures applied to the sediment samples, as reported in a recent study (Ciuffo et al., 2002). However,

only a few reports have appeared on the nature of such relationships in marine sediments.

4.2.3 ADSORPTION

In the hydrosphere, the prevailing aqueous species of cesium is the uncomplexed Cs^+ ion, and changes of pH and Eh do not affect the speciation of cesium (Forsberg et al., 2000). Nonetheless, cesium may be adsorbed onto the surfaces of colloids and suspended particles that deposit onto aquatic system bottoms. The ability of bottom sediments to bind cesium varies with particle size, and an increase in ^{137}Cs activity in sediment with a decrease in particle size has been observed (Forsberg et al., 2001). According to Lujanien et al., the highest calculated K_d values ($K_d 2 \times 10^3$; i.e., the equilibrium ratio of ^{137}Cs in particles compared with the ^{137}Cs in the water phase) were obtained for particles smaller than 4 mm (Lujanien et al., 2005; Kruyts and Delvaux, 2002). The reason for this observation may be because the high adsorption of Cs^+ ions is mainly determined by the clay minerals present in the sediments (Kruyts et al., 2004). The main mechanism of the adsorption is ion exchange, and in natural sediments, the sorption behavior is dominated by the highly selective exchange sites in clay minerals (Nikolova et al., 2000). The adsorption has been reported as virtually irreversible (Catalano et al., 2006). Although cations with similar charge and ionic radii are expected to compete with cesium for the adsorption sites, the bonding strength observed for clay minerals decreases in the sequence $\text{Cs}^+ > \text{Rb}^+ > \text{K}^+ > \text{Na}^+$ (Chen et al., 2005), which is in agreement with the decrease in ionic radius and the increase in the hydration enthalpies.

The single most important property affecting the characteristic of transuranium elements is their oxidation state. The precipitation, complexation and sorption processes depend on the prevailing oxidation state (Chorover et al., 2003; Chorover et al., 2008). All four processes contribute to the chemical behavior and environmental transport properties of the actinides in the environment, including plutonium and americium.

In the clay mineral muscovite, a negative fixed charge arises primarily from isomorphous substitution of Al^{3+} for Si^{4+} in the tetrahedral sheet comprising the siloxane site (Liu et al., 2006). In the illite mineral, whose composition is quite close to that of muscovite, the isomorphous substitution of Al^{3+} for Si^{4+} and partial substitution of Fe^{2+} and Mg^{2+} for Al^{3+} enhances the stability of the Cs^+ -siloxane surface complex, presumably by promoting the dehydration of the adsorbed Cs cation (Mon et al., 2005). The layer-type silicates bind Cs either through weak electrostatic interactions or through stronger bonds formed by partial sharing of electrons between Cs and the ligand sites of the clay mineral (Hakanson, 2000). The electrostatic associations of the hydrated Cs with anionic surfaces within the basal plane or interlayer and the dissociated edge hydroxyl groups act as outer-sphere complexes (Pinder et al., 2010; Smith, 2006). Electronic bonding at the frayed

edge sites or external basalt sites or within the interlayer leads to inner-sphere complexes, which usually are much more stable than the outer-sphere complexes. Cesium adsorbed to outer-sphere complexes can be easily desorbed and is more mobile in the environment, whereas inner-sphere sorption complexes may limit the Cs mobility and bioavailability (Mon et al., 2005). These inner-sphere adsorption complexes are most likely the dominant species at low concentrations typical of radioactive Cs contamination due to the negative charge and strong hydrophilicity, and a great amount of Cs is expected to be strongly adsorbed from contaminated environments.

The complicated behavior of cesium may be a result of peculiarities of its sorption to various sites of different clay minerals and their mixtures. Radiocesium materials in aqueous environment can occur as solids, colloids, or solvated species (Smith et al., 2006). The presence of these species is regulated by thermodynamic and kinetic laws and is sensitive to such parameters as cation–anion concentration, ionic strength, temperature, gas–liquid–solid phase equilibria, and oxidation–reduction potential (International Atomic Energy Association, 2009). The complicated behavior of cesium in the environment can be generally explained by its sorption to various sites of different silicates. In smectites, the isomorphous substitution in both the tetrahedral and octahedral layers generates weak negative charges of the sheets, thus leading to the formation of unstable structures and resulting in wide ranges of cation exchange capacity, selectivity and swelling properties (Cook et al., 2007). Water and such cations as H^+ , Na^+ , Ca^{2+} , and Mg^{2+} can easily penetrate into the smectite interlayer and participate in exchange processes (Smith et al., 2006). Mixtures of clay minerals (e.g., illite and smectite) present in the natural environment can cause difficulties in the quantitative estimation of exchangeable and bioavailable cesium; therefore, semi-empirical methods are used for these purposes (Cook et al., 2007; International Atomic Energy Association, 2009). Unlike other heavy metals, Cs does not have an affinity to sorb onto or co-precipitate with Fe-hydroxides. However, organic matter and iron oxides have indirect effects on the ^{137}Cs affinity for the clay minerals (International Atomic Energy Association, 2004). The adsorption of organic molecules on clay minerals influences their affinity to cesium, and the clay-humic substance complexes adsorb a greater amount of cesium than uncoated clay minerals (Fisher et al., 1999). Coatings of iron oxides block the cesium uptake sites as well. The organic matter and iron oxide coatings on soil and sediment particles can serve as intermediate phases when the exchangeable ^{137}Cs permeates toward the mineral core of clay particles. Moreover, the desorption of exchangeable ^{137}Cs from clay minerals can be inhibited by such coatings. As a result, the value of the exchangeable ^{137}Cs measured during the extraction procedures can be reduced. In addition, organic matter enhances the sorption and “fixation” of Cs^+ ions in the clay interlayer (Fisher et al., 1999). However, the mechanisms of these interactions are poorly understood.

^{137}Cs has a high affinity for suspended particulate matter, especially for micaceous clay minerals (Butkus et al., 2009). This process leads to the rapid removal of ^{137}Cs from the water column (Forsberg et al., 2000) and a notably long residence time in catchment soils (Ciuffo et al., 2002). Although it occurs rapidly, the adsorption is reversible in the presence of competing cations, particularly NH_4^{4+} , which is usually abundant in profundal lake sediments (Cook et al., 2007; International Atomic Energy Association, 2009; Smith et al., 2006). Thus, surface waters continue to receive ^{137}Cs through internal loading from sediments in addition to the continual slow flux from catchment soils.

4.4 Distribution of ^{137}Cs in the Biotic Components of Aquatic Ecosystems

In addition to the abiotic processes described previously, Cs+ recycling and mobilization from sediments may also be mediated via biotic processes. Marine biota concentrate the radionuclides in their systems through direct absorption and/or via the ingestion of other organisms and form fecal pellets, which subsequently accelerate the sinking of radionuclides (Bellenger et al., 2008). The association of Cs^+ with suspended plankton in the water column can substantially increase the retention time of Cs in water (Bostick et al., 2002), and microbial activity has been implicated in the mobilization of Cs^+ from sediments (Cha et al., 2006). Furthermore, filter feeding fish may accumulate Cs adsorbed to particulate matter, and benthic invertebrates may become associated with contaminated sediments (Cremers et al., 1988). The remobilization of particle-bound Cs by these organisms is of particular importance in aquatic ecosystems, in which Cs^+ may be transferred along the food chains with high efficiency. Pendleton (1960) initially demonstrated that the ^{137}Cs in freshwater ponds becomes concentrated in progressively higher levels as it is transferred up the food web via bioconcentration or bioaccumulation, and it is now well established that most ^{137}Cs accumulated by higher organisms (e.g., fish) in aquatic habitats originates from the ingestion of ^{137}Cs -contaminated organisms rather than from the direct absorption of the radionuclide from water (Fan, 2012; Giannakopoulou et al., 2007; Honda et al., 2012). In addition, the potential role of microbial primary producers (e.g., cyanobacteria and microalgae) in the mobilization and cycling of cesium in aquatic ecosystems has been recently highlighted (Kato et al., 2012; Tanaka et al., 2012).

4.4.1 PHYTOPLANKTON

Aquatic plants may obtain nutrients from the sediments by root uptake or may absorb nutrients from the water column by foliar uptake (Nikolova et al., 2000; Seaman et al., 2001). Because the ^{137}Cs in rivers is rapidly incorporated into sediments, it may be expected that root uptake would be

the principal mechanism for ^{137}Cs uptake by water plants rooted in the sediment. In certain cases, plants such as *Cladophora glomerata*, *M. spicatum*, *Potamogeton pectinatus*, and *Ceratophyllum demersum* absorb cesium or potassium directly from the water column (Camps et al., 2003). Foliar absorption must be considered as a potential mode of ^{137}Cs accumulation in water plants as well (Xiong et al., 2005). Similar morphological peculiarities of the submerged perennial plant *M. spicatum* (milfoil) with long, branching stems and feather-like leaves may be the reason for their high ^{137}Cs content. However, *Potamogeton nodosus* (pondweed), with relatively large, floating leaves, has a limited surface area to biomass ratio. The lower value of ^{137}Cs content in the floating leaves may be explained by the fact that only their undersides are exposed to the water environment (50% of the total surface; Henley et al., 2000). In general, submerged plants (*Cladophora glomerata*, *M. spicatum*, *Potamogeton pectinatus*, *Ceratophyllum demersum*) have higher levels of ^{137}Cs contamination than semi-aquatic plants (*Cyperus longus* or *Paspalum pasalodes*). The fact that both of the previously mentioned plants (*Ceratophyllum demersum* and *M. spicatum*) are longer-lived, perennial species gives them the opportunity to accumulate radionuclides over a longer period and hence to a greater degree. Other rooted (or at least anchored in the sediments) macrophyte species, such as *Cyperus longus* and *Paspalum pasalodes*, may absorb nutrients from either the water column or the sediments (Nikolova et al., 2000). These species may affect the relative absorption of nutrients from both water and sediments. In the case of *Cyperus longus*, the roots revealed higher ^{137}Cs concentrations than the leaves, whereas the stems showed the lowest concentrations. Mineeva et al. (1990) used *Lamium maculatum* as an example and also reported that in aquatic plants, the greatest concentrations occur in the root system, with the coefficient in the roots measured at values 1.5 times higher than that of the aboveground parts (Camps et al., 2003). One of the reasons for this observation may be the adherence of sediment particles to the roots.

Different species of aquatic plants have different affinities for ^{137}Cs or potassium. The differences in ^{137}Cs values may be attributed to the relationship between ^{137}Cs (or potassium) uptake and the metabolic status of plant tissues in terms of chlorophyll, sugar or starch concentrations (Xiong et al., 2005). The stem, which exhibits lower physiological activity than the leaves, consists of vascular tissue and accumulates ^{137}Cs (similar to potassium) to a lesser degree than the leaves. Furthermore, the leaves of aquatic plants (especially the submerged plants *Ceratophyllum demersum* and *M. spicatum*) are extensively influenced by the aquatic environment in comparison with terrestrial plants (Camps et al., 2003). This observation may reflect competition between the K^+ , Ca^{2+} , Mg^{2+} , and Fe^{2+} elements for binding or for uptake sites in the aquatic plants (Nikolova et al., 2000). External Ca^{2+} or K^+ concentration is an important control variable because the ^{137}Cs accumulation in plants can be largely affected (Henley et al., 2000). The large

values indicated in the submerged free-floating *Ceratophyllum demersum* (coontail) may be attributed to the division of the leaves into many narrow and tapering segments. The large surface area increases their exposure to the water environment and thus their uptake ability. The rather stiff and tapering leaf segments and the small teeth along the margin increase the possibility of trapping sediment particles suspended in the water column. Apart from physical levels or plant morphology, the ^{137}Cs uptake is influenced by allometric parameters and may be regulated by physiological mechanisms, as is the case for heavy metal accumulation (Yasunari et al., 2011).

The various growth forms of aquatic macrophytes present opportunities and impose constraints on nutrient uptake. For free-floating plants such as duckweeds (*Lemna spp.*), whose roots are suspended in the water column, the source of nutrient uptake is constrained to the water column. These species occur in three primary growth forms (i.e., emergent, floating-leaf, and submerged), and these forms may affect the relative absorption of nutrients from water and sediments (Nikolova et al., 2000). Emergent species such as cattails (*Typha spp.*) are rooted in the sediments of primarily shallow water and have erect stems that project above the water surface (Xiong et al., 2005). Floating-leaf species such as water lilies (*Nymphaea spp.*) are rooted in (or are at least attached to) the sediments and have stems or petioles that extend to floating leaves. Submerged species, such as the familiar aquarium plant elodea (*Elodea spp.*), are rooted in deeper waters and have stems and leaves that extend upward into the water column (Yasunari et al., 2011). For most submerged species, the uptake of nitrogen and phosphorus appears to occur from the sediments, but at least a portion of the uptake of cations occurs by foliar absorption from the water column (Bunzl, 2002). Foliar absorption of potassium has been either demonstrated or indicated for submerged macrophytes (Butkus et al., 2009). Foliar absorption of sodium (Na) and rubidium (Rb) has been also demonstrated by Jacoby et al. (1973) and Waisel et al. (1982), but foliar absorption of cesium has not been experimentally demonstrated for submerged macrophytes (Ciuffo et al., 2002).

Little evidence exists for the foliar absorption of cations by floating-leaf species. Foliar uptake of ^{137}Cs has been demonstrated for the floating-leaf species *Brasenia schreberi*, *Nymphaea odorata*, and *Nymphoides cordata* (Yasunari et al., 2011). Emergent species commonly have extensive root systems, and only a portion of their foliar mass is immersed in the water column; consequently, root uptake is presumed to be the dominant mechanism of absorption (Nikolova et al., 2000). However, evidence indicates that the foliar uptake of Cs does occur in emergent species. Myttenaere et al. (1969) showed that grain absorbed more ^{137}Cs from the water than from the roots (Camps et al., 2003). The results of Pendleton (1960) suggest foliar uptake of ^{137}Cs in the emergent macrophyte *Polygonum persicaria* (Butkus et al., 2009). Most of these studies of foliar absorption have been performed on relatively small scales, and numerous studies have used excised tissues in laboratory cultures

(Tanaka et al., 2012) or whole plants in either aquarium- (Ciuffo et al., 2002) or mesocosm-sized (Forsberg et al., 2000) facilities. Certain studies have used limited-scale manipulations or transplants within whole-lake systems (Ciuffo et al., 2002). These studies have been characterized by mostly qualitative demonstrations that foliar uptake occurred and were designed to address the more physiological question of how formerly terrestrial angiosperms have adapted to varying levels of immersion in aquatic habitats.

In the cases in which radioactive isotopes are involved, more practical questions arise, such as how the rates of foliar absorption can affect the transport and fate of potential releases to aquatic systems. The isotopes of Cs may be of special concern for the following reasons: (a) their similar behavior to the required element K in plants; (b) their solubility in aquatic systems; (c) the vaporization, release, and dispersal of ^{134}Cs and ^{137}Cs in major reactor accidents (Forsberg et al., 2001); and (d) the abundance and persistence of ^{137}Cs in spent fuel and reprocessed wastes (Butkus et al., 2009). The foliar absorption rates measured using the techniques of the previous studies, although these techniques were appropriate for the purposes of those studies, may have limited ability to predict the transport and fate of Cs and other elements at the scales of ponds, lakes, and reservoirs. The rate of Cs absorption by macrophyte species could be determined by measuring the changes in plant Cs concentrations following the experimental or accidental release of Cs isotopes into the water column. However, most studies of these releases do not report sufficient analyses of macrophyte concentrations to assess the foliar absorption, and those with sufficient data have not specifically analyzed the data to quantify the potential absorption processes. The possible presence of foliar absorption of Cs (and other elements with important radioactive isotopes) and the absence of appropriate absorption rate estimates for those elements suggest potential limitations in the ability to assess the transport and fate of accidental releases where macrophytes are abundant. For example, the ^{137}Cs inventory in the foliar-absorbing species *B. schreberi*, *N. odorata*, and *N. cordata* can be as large as that in a pond's water column (Ciuffo et al., 2002; Forsberg et al., 2000), but little is known about how these species affect the initial transport and fate of accidental releases.

The sediment acts as a sink for ^{137}Cs ; thus, the availability of the nuclide for biological incorporation by plants is limited. Plants act as an intermediate in the migration of ^{137}Cs in an aquatic ecosystem because the half-life of ^{137}Cs is much longer than the lifetime of the plants. Thus, it is necessary to estimate the retention of radionuclides by the plant biomass and the release of ^{137}Cs from the biomass back into the water and sediment, where it can become biologically available once again (Forsberg et al., 2001). Certain species that are highly efficient in the uptake and retention of ^{137}Cs can be used as environmental bio-indicators for rough estimates of the effective dose to certain individuals (Kruyts and Delvaux, 2002). Carignan (1985)

TABLE 3. Species for which the foliar uptake of alkali elements has been experimentally demonstrated

Serial no.	Macrophyte Species	Elements	Sources
Submerged Species			
1	<i>Callitriche bamulata</i>	K	(Adamec, 1997)
2	<i>Elodea canadensis</i>	K	(Adamec, 1997)
3	<i>Hydrilla verticillata</i>	K	(Barko, 1982)
4	<i>Myriophyllum heterophyllum</i>	K	(Mickle and Wetzel, 1978)
5	<i>Myriophyllum spicatum</i>	K, Na, Rb, Cs	(Carignan, 1985; Pinder et al., 2006; Waisel et al., 1982)
6	<i>Najas marina</i>	Na, Rb	(Waisel et al., 1982)
7	<i>Potamogeton pectinatus</i>	K	(Fiol et al., 2005)
8	<i>Potamogeton perfoliatus</i>	Na, Rb	(Waisel et al., 1982)
9	<i>Potamogeton lucens</i>	Na, Rb	(Waisel et al., 1982)
10	<i>Ranunculus aquatilis</i>	K	(Adamec, 1997)
11	<i>Ranunculus trichophyllum</i>	K	(Adamec, 1997)
12	<i>Scirpus subterminalis</i>	K	(Mickle and Wetzel, 1978)
13	<i>Utricularia inflata</i>	Cs	(Pinder et al., 2006)
14	<i>Vallisneria spiralis</i>	Rb	(Pinder et al., 2006)
Floating leaf species			
15	<i>Brasenia schreberi</i>	Cs	(Kelly and Pinder, 1996; Pinder et al., 2006)
16	<i>Nymphaea odorata</i>	Cs	(Kelly and Pinder, 1996; Pinder et al., 2006)
17	<i>Nymphoides cordata</i>	Cs	(Kelly and Pinder, 1996)
Emergent species			
18	<i>Oryza fatua</i>	Cs	(Myttenaere et al., 1969)
19	<i>Polygonum persicaria</i>	Cs	(Pendleton, 1960)

suggested that *M. spicatum* transports K^+ absorbed from the water column to the roots, where it is released into the sediments to displace NH_4^{4+} that can augment N availability; a similar process occurs for Cs, and a portion of what appears to be loss may instead be translocation to roots. For the emergent species rooted in the sediments, there was little evidence of foliar uptake. The lack of a clear indication of foliar uptake in *S. latifolia* and *T. latifolia* is consistent with the evidence from other studies showing the dependence of K and Cs concentrations on sediment properties (Kruyts et al., 2004; Nikolova et al., 2000). Foliar uptake of alkali elements has been demonstrated or indicated for macrophyte species (Table 3). Although certain species, such as *Callitriche bamulata* (Ciuffo et al., 2002) and *Scirpus subterminalis* (Forsberg et al., 2000), appear to indicate less absorption than others, most species show some indication of foliar uptake. Reports showing the absence of foliar uptake in submerged or floating-leaf species appear to be lacking. It would thus seem reasonable to expect a certain degree of foliar uptake of Cs isotopes in most aquatic systems with either submerged or floating-leaf species. Although foliar uptake has been commonly demonstrated for aquatic macrophytes, most models of the transport and fate of Cs in lakes

(Chen et al., 2005; Chorover et al., 2003; Chorover et al., 2008; Liu et al., 2006) do not contain components and pathways that consider macrophytes. However, these models have been accurate predictors of Cs concentrations in water and fishes (Hakanson, 2000; Mon et al., 2005).

Whether surface or internal, the Cs associated with these macrophytes is retained in the biotic system and is lost in outflow from the lake or sequestered by the sediments. It remains available for entry into the food chains through either the ingestion of whole tissues or, if surface deposited, through surface cleansing by such organisms as gastropods. Perhaps the most important questions from a transport and fate perspective include what proportions of this Cs are (a) sequestered in the sediments by translocation to and release from roots, (b) incorporated at the surfaces of littoral sediments after release from decomposing plant tissues, or (3c) returned to the water column (Pinder et al., 2010).

4.4.2 ZOOPLANKTON

The ability of aquatic organisms to accumulate Cs^+ is markedly influenced by salinity. The higher cation content of seawater than freshwater results in ^{137}Cs concentration factors of marine organisms that are several times lower than those generally observed in freshwater organisms (Smith, 2006; Smith et al., 2006). This condition may be reflected in levels of ^{137}Cs in humans for whom fish is an important component of the diet. Individuals who feed on freshwater fish may acquire ^{137}Cs burdens that are several times higher than those of individuals feeding on marine fish or more diversified diets (Cook et al., 2007; International Atomic Energy Association, 2009). However, it should be noted that in certain cases, the presence of Na^+ might result in the enhanced accumulation of Cs^+ by aquatic organisms. For example, the Cs^+ influx in the estuarine green microalga *Chlorella salina* via a K^+ uptake mechanism employed to counteract hyper-osmotic shock was increased several-fold at elevated NaCl concentrations (Pinder et al., 2006). Although the Na^+ ions can inhibit Cs^+ accumulation in most organisms, it is the higher concentration of K^+ in water that is generally considered to exert the greater inhibitory effect on Cs^+ accumulation in aquatic organisms. In freshwater habitats, increased Cs/K ratios are also correlated with increased Cs^+ uptake by fish (International Atomic Energy Association, 2009).

Svadlenkova et al. (1989) modeled ^{137}Cs uptake by the alga *Cladophora* and estimated that a 50% increase in the K^+ concentration of water results in a 30% decline in Cs^+ uptake, whereas a 50% reduction in the K^+ concentration results in a 100% increase in Cs^+ uptake levels (Tang et al., 2003). The application of the model proposed by the authors may have potential for the use of algae as bio-indicators of aquatic ^{137}Cs levels. It should be noted that the uptake of Cs^+ under conditions of K^+ -deficient waters may be beneficial to certain organisms. For example, it has been demonstrated that the respiration of the marine alga *Porphyra perforata* starved of K^+ may

be partially restored in the presence of Cs^+ (Tsukada et al., 2002). The retention time of Cs^+ also markedly affects the Cs^+ accumulation by aquatic organisms. Clearly, the longer an organism is exposed to Cs^+ , the greater the likelihood that Cs^+ uptake will occur.

Indeed, it has been suggested that, apart from the initial level of fall-out entering aquatic systems, the individual factor of greatest importance in determining ^{137}Cs concentration factors of freshwater fish following the Chernobyl accident has been the water retention time of ^{137}Cs in rivers or lakes (Tang et al., 2003). Fish in lakes may take in up to 10 times more Cs^+ than fish living in flowing rivers (Pinder et al., 2006). However, because of the continued release of Cs from surrounding drainage areas, the net rate of elimination of Chernobyl-derived ^{137}Cs from samples of water, sediment, algae, and fish in the rivers is low. Thus, even in the most mobile aquatic habitats (i.e., flowing rivers), Cs^+ may persist in a biologically available form for several years after release.

The hypothesis concerning the impact of pH on the transfer of ^{137}Cs into fish was also presented by Tsukada et al. (2002). The effective uptake and long retention time of ^{137}Cs in fish are linked with the oligotrophic character of the aquatic systems associated with a deficiency of potassium in water. It is well known that ^{137}Cs is transferred much more effectively into fish in oligotrophic lakes than in eutrophic lakes (Pinder et al., 2006; Sawidis et al., 2003; Tang et al., 2003; Tsukada et al., 2002). Särkkä et al. (1990) noticed that small values of total phosphorus concentration, conductivity, color, and pH tended to increase the cesium content in fish (Strezov et al., 2009). Furthermore, Särkkä et al. (1996) stated that water color has no effect on the sedimentation of cesium, unlike the effect observed on fish (Strezov and Nonova, 2005a). Penttilä et al. (1993) proved that humic substances wield only a small influence on the bioavailability of ^{137}Cs in lake water (Burger et al., 2006). Potassium, a chemical analog of cesium, exists in solution in ionic form and will not form inorganic complexes (Skwarzec et al., 2003). Särkkä et al. (1996) found that all of the water quality parameters in their study were positively correlated with the activity concentration of ^{137}Cs in fish, but the activity concentrations of ^{137}Cs in bottom sediments were not (Ciuffo et al., 2002). The ^{137}Cs concentration in fish showed a positive correlation with the original deposition of ^{137}Cs and water residence time and a negative correlation with the size of the catchment area, phosphorus content, water color, pH, conductivity of water and potassium content in water. These authors also stated that higher activity concentrations were noted in lakes with less humic substances than in lakes with an abundance of humic substances. In contrast, Sonesten (2000) found that the amount of ^{137}Cs in fish correlated positively with the amount of humic substances in lake water (Tanaka et al., 2012).

Water color is inversely correlated and pH is positively correlated with the bioconcentration factor of plankton (Tang et al., 2003). Among other

components, Särkkä et al. (1990) reported a negative correlation between the original deposition of ^{137}Cs and the water color, pH, conductivity, and potassium and phosphorus content of lake water. The higher transfer of ^{137}Cs into large perch may result from changes in the feeding habits of *Perca flavescens* as they grow, beginning with plankton and invertebrates and progressing to small fish. Different types of lakes show different behaviors for the transfer of ^{137}Cs in different fish species due to variations in the lake limnology and the nature of their catchments. In fish, ^{137}Cs activities also vary considerably as a function of both size and species (Skwarzec et al., 2003). A certain amount of variation may also be due to the selective sequestration of Cs^+ in particular organs (e.g., Cs^+) may become more readily incorporated into soft tissues (muscle, liver) than into bones (Stengel et al., 2004). Durand et al. (1994) presented a detailed characterization of Cs^+ transport and fate in freshwater and marine organisms (Strezov and Nonova, 2005b). The authors found that Cs^+ behaved as a soluble ionic component of the cytosolic fractions in lobsters, oysters, and eels, with no detectable association of Cs with high or intermediate molecular weight proteins.

The dependence of the transfer of ^{137}Cs into fish on water chemistry, however, has been widely studied. Rowan and Rasmussen (1994) reported a model describing a relationship between the concentration factor, the trophic status of the fish (piscivorous, omnivorous, and median), the amount of suspended solids, and the potassium content of lake water (Skwarzec et al., 2003). The inverse correlation of the transfer of ^{137}Cs into fish with the potassium content of water was also found in earlier studies (Strezov and Nonova, 2005a; Varskog, 1994). Although Cs is not an essential element, it enters food webs through the K channels in plant membranes and is subsequently transferred to consumers through ingestion. Aquatic consumers acquire only a small fraction of their ^{137}Cs through direct uptake from water. Early studies on a variety of aquatic organisms, ranging from plants and invertebrates to fish, showed tissue/water bioconcentration factors of 2–4 orders of magnitude. In a recent review of radiocesium bioaccumulation by fish (Kelly and Pinder, 1996), the uptake of radiocesium in fish was shown to decrease with increasing K and suspended sediment concentrations in water and approximately doubled with each trophic level in the food web. These empirical models have been adopted by the International Atomic Energy Association (1994) for use in risk assessment and are highly successful in explaining the variation within and between systems; however, they do not explain why two-fold differences often exist within a species or between species at the same trophic level within a system.

The studies discussed in this work indicate that the bioaccumulation of Cs^+ from the environment occurs readily in most groups of organisms, although uptake levels vary considerably, and a strong inhibition of uptake may result in the presence of other monovalent cations. Organisms occupying low trophic levels (e.g., micro-organisms, phytoplankton, higher

fungi, mosses and plants) appear to be primarily responsible for the direct removal of radiocesium from the environment. Subsequent transfer through the food chains is likely to be the most important route of Cs^+ accumulation in higher organisms (e.g., cattle and humans; additional Cs contamination may arise through the inhalation/absorption of Cs from their immediate environment as well as the incidental consumption of soil/sediment particles). Thus, Cs^+ accumulation in humans can be strongly influenced by the nature of the human diet. The tendency of Cs^+ to remain in soluble form in aquatic ecosystems predisposes humans to contamination when sea and freshwater (lower concentrations of K^+ and Na^+) food form an important component of the diet.

5. MEASURES FOR REDUCING THE RISK OF ^{137}CS IN AN AQUATIC ECOSYSTEM

^{137}Cs is the radionuclide that generally drives human health and ecological risks in the ecosystem, and thus it is subject to remediation operations. Aquatic ecosystems, including lakes, rivers, groundwater and seas, all have site-specific behavior governed by the hydrological and morphological parameters of the water bodies and their drainage areas. The remediation of such water bodies following the contamination of ^{137}Cs is therefore largely dependent on these site-specific parameters. In addition, general remediation plans for contaminated waters may be expensive and may include large engineering costs. The effects of such remediation efforts therefore must normally occur on a cost-benefit basis and should be chosen according to the well-known ALARA (as low as reasonably achievable) principles and include comparison with risks from other toxic substances present in the water. The remediation of a contaminated ecosystem involves physical, chemical, and biological procedures, which are described subsequently.

5.1 Biological Remediation

This section addresses the use of micro-organisms and aquatic plants to alter the distribution or mobility of ^{137}Cs . Micro-organisms can alter the mobility of ^{137}Cs but cannot be directly removed as can aquatic plants (Eapen, 2004). Phytoremediation includes phytoextraction (the concentration of harvestable portions of plant biomass) and phytostabilization (the use of plants to minimize off-site losses of ^{137}Cs through erosion and leaching). Factors such as the cleanup goals, level of radionuclide contamination, level of contamination, water chemistry, presence of other toxic materials, disposal of harvested biomass, and climatic conditions can all influence the likelihood of success (Smolders et al., 2011). Phytostabilization may already act as an important process at certain sites wherever natural vegetation intercepts the ^{137}Cs migrating from contaminated sites to the surface waters.

Phytostabilization and phytoextraction can have equally important but different roles in the phytoremediation of sediments contaminated by ^{137}Cs (Cobbett, 2000).

5.1.1 PHYTOREMEDIATION

Phytoremediation has been proposed as a promising alternative; it is an eco-friendly, cost-effective, in situ treatment technology that relies on the capacity of plants and their associated micro-organisms to stabilize or extract contaminants from aquatic ecosystems. The potential of *C. roseus* plants has been studied for the uptake of ^{137}Cs at three different activity concentrations in sediments and has been found to remove 73%, 59.3%, and 51.3% of ^{137}Cs within 15 days of the experiment, respectively (Radway et al., 2001). The research findings proved that ^{137}Cs and ^{90}Sr are bio-accumulated by this weed plant, which may be an ideal hyper-accumulator with a high potential for the removal of radionuclides. After remediation, this weed can be harvested, burned to ash, and disposed of as waste in a safe environment. The aquatic plant *Lemna gibba* was used to biosorb ^{137}Cs from aqueous radioactive waste simulate (Kleinschmidt, 2009). A study using batchwise laboratory-scale experiments demonstrates that the parameters that may affect the efficiency of *Lemna gibba* in the bio-removal and bioaccumulation of the two radionuclides include the contact time, the pH value and the initial activity content of the waste simulate in addition to the light effect and amount of biomass. The uptake values, biosorption efficiency percentages, rate constants, and isotherm factors were evaluated for this process. The respective uptake values from waste-simulate solution containing 6100 Bq for ^{60}Co and ^{137}Cs at $\text{pH} = 6.9$ after 24 hr contact were recorded as 1213 Bq/gm and 872 Bq/gm (Kruyts et al., 2004). The results obtained show the potential of the aquatic plant *Lemna gibba* for use as an efficient biological sorber for ^{137}Cs from streams with low and intermediate levels of aqueous radioactive waste.

5.1.2 WETLANDS

Aquatic plants can also be used for wetland development. The most important role of plants in wetlands is that they increase the residence time of water and thereby increase the sedimentation of particles and associated pollutants (Cobbett, 2000). Thus, these plants are indirectly involved in the cleaning of water. Plants also add oxygen to the roots, generating favorable conditions for microbes and bioremediation. For the efficient removal of pollutants, a high biomass per volume of water of the submerged plants is necessary (sukada et al., 2002), and uptake of metals in emergent plants only accounts for 5% or less of the total removal capacity in wetlands.

5.1.3 BIOLOGICAL DILUTION

To reduce the bioavailable amount of ^{137}Cs in the lake, a method known as biological dilution can be used. The intent is to remove ^{137}Cs already taken

up in the food web by reducing the fish stock (intensive fishing) in the lake and to influence the predation pressure such that the food web is shifted and will contain a relatively larger amount of plankton, which may result in lower concentrations of ^{137}Cs in the biota by means of biological dilution to decrease the concentration of ^{137}Cs in each individual fish (Schwaiger et al., 2004). For this purpose, the lakes can be treated with different types of fertilizer (discharges from aquaculture, commercial fertilizer, and P-enriched lime; Franić, 2008). The goal is to increase the lake biomass and thereby disperse the given amount of ^{137}Cs (Yamada and Nagaya, 1998). The method is based on theories of biological buffering.

5.2 Chemical Remediation

A few chemical remediation techniques currently exist and are available for ^{137}Cs . Dodge and Francis (1994) developed a process to recover ^{137}Cs from soils using citric acid and visible light photodegradation (Morita, 2005). Their early studies have shown that the uranyl ion is photochemically active in the presence of organic acids, and upon exposure to visible light, the uranyl citrate complex undergoes photochemical oxidation/reduction reactions (Koulikov, 2003). The mixture of citric acid and contaminated soil is first treated with bacteria, which will degrade the complexed citric acid to carbon dioxide and water. The supernatant containing the uranium-citrate complex is subsequently separated and subjected to photodegradation for uranium recovery (Fesenko, 2010). It is doubtful, however, that such approaches would aid ^{137}Cs extraction from contaminated sediments.

5.2.1 FERTILIZERS

Chemical amendments in the form of fertilization practices have been shown to have dramatic effects on plant accumulation of ^{137}Cs and thus on the effectiveness of phytoextraction. The most important nutrients in the aquatic ecosystems are phosphorus and nitrogen (Smith et al., 2002). Total phosphorus has long been recognized as the nutrient most likely to limit primary lake productivity. Because the most important focus of the remediation of aquatic ecosystems is the adaptation of potassium (K) and nitrogen (N) fertilization techniques, management practices for land use and agricultural water may be the best options for minimizing radioactive ^{137}Cs in the local food chain in natural ecosystems (Sundbom, 2003). In particular, information on the K status of the ecosystem will be essential to predicting the efficiency of K fertilizer application in reducing the transfer of ^{137}Cs . Several practical and ecologically relevant methods exist to remediate lakes contaminated by ^{137}Cs , including liming, potash treatment and the fertilization of low-productive lakes (Tostowaryk, 2000).

Nitrogen fertilization also results in increased ^{137}Cs uptake by increasing plant growth rates, whereas fertilization with large amounts of potassium results in a substantial reduction in ^{137}Cs uptake (Franić, 2008). Ammonium

sulfate fertilizers can be also used for growth stimulation and to displace the exchangeable fraction of ^{137}Cs in the soil matrix with the ammonium ion, thus increasing the uptake availability of ^{137}Cs . Those fertilizers containing competing elements (e.g., potassium) will have significant effects on the ^{137}Cs behavior and plant-root uptake (Sysoeva, 2005). Potassium fertilization has been shown as a successful counter measure for ryegrass until the potassium loading in soil reaches $\sim 5\%$ of the cation exchange capacity (CEC). Above this value, the soil-to-plant transfer may increase as the potassium supply increases because the K_d decreases, and the concentration ratio may remain essentially constant (Hattink, 2009). In sandy soils, especially those with generally low CEC values and low K_d values, only a small range of potassium fertilization levels can be used as a counter measure for ryegrass. In certain instances (e.g., with spinach), the correlation between the soil-to-plant transfer and the K_d is such that potassium fertilization would not necessarily act as a successful counter-measure (Evans, 2005).

5.2.2 POTASH TREATMENT

Blocking and/or substitution can reduce the proportion of ^{137}Cs taken up by fish. For this reason, potash treatment has been carried out to increase the potassium concentration of lake waters. Potassium and cesium are taken up in fish in a similar manner (Tyler, 2001), as described previously. Other ions may also participate in different blocking processes (e.g., Ca, Mg, and Na), which implies that the different liming measures, which produce a general increase in the ionic strength of the water, may also produce a positive effect in this way. However, one can increase pH without changing the concentration of potassium in the lake by adding primary rock lime with no potassium, but it is not possible to increase the K concentration without increasing the lake pH at the same time.

5.2.3 LIMING

The process of lake liming has been carried out to increase the proportion of ^{137}Cs sedimented on the lake bottom and thereby prevent or delay its bio-uptake. The hypothesis is that the flocculation tendency of the ^{137}Cs -carrying particles is increased by increasing the pH, alkalinity and hardness of the lake water (Hann et al., 2001). Naturally low pH occurs in many oligotrophic lakes with catchments dominated by acidic rocks and mires, and many natural processes and properties in the catchment influence the lake pH. This scenario means that in the natural, preindustrial, or pre-civilization state, the pH varies from lake to lake. It is clearly not possible in the present to measure the conditions of the past, but predictive methods are available. In Sweden, a crude rule-of-thumb system is applied by the National Environmental Protection Agency and regional authorities. In which lakes are generally limed to approximately 6.4–6.5 (Møller, 2006). The natural range

of the mean annual pH in small glacial lakes would vary from approximately 5.5 to approximately 7.2, and the value 6.4 is only occasionally correct.

5.2.4 AMMONIUM ADDITION

Recent studies show that the addition of ammonium increases the availability of ^{137}Cs for plants growing in aged contaminated soils (Hesse-Honegger and Wallimann, 2008). Field trials associated with this research indicate that plants that have both a high uptake and high biomass production could be used to remediate a ^{137}Cs contaminated site (Marmioli, 2003). Historical research also has shown that the addition of ammonium-based fertilizers increases ^{137}Cs uptake. Thus, there appears to be a certain potential for enhancing the plant uptake of soil ^{137}Cs by the application of ammonium fertilizers (McCutcheon, 2003). Changes in soil chemistry caused by rhizosphere development may subsequently promote or inhibit plant uptake of radionuclides, but these changes are not yet fully understood.

5.3 Physical Remediation

A number of different physical methods exist for stabilizing, immobilizing or removing contaminants in aquatic ecosystems. Physical processes generally apply to a wide variety of contaminants in various chemical forms. The physical remediation of radioactive constituents does not alter the level of radioactivity unless the contaminated soil or sediment is physically removed but may reduce the potential for exposure or movement through the soil or sediments.

5.3.1 CAPPING AND IN SITU GROUTING

Because ^{137}Cs binds tightly to clays, capping and in situ grouting that prevent water intrusion and solution phase migration of contaminants are generally not as cost effective as other methods (Singh, 2008). Sulfur polymer cement, saltstone, and solidification are a subset of the methods for containing wastes in cementitious forms. Electrokinetic techniques require that the ionic species under remediation must be dissolved in the soil water to facilitate movement to a collecting electrode (Singh, 2009). A large fraction of ^{137}Cs , however, will likely become trapped in the soil structure and not in the soil solution.

5.3.2 ADDITION OF MICACEOUS MINERALS

The high ^{137}Cs mobility in aquatic systems can be due to the low retention capacity of the kaolinitic dominated sediments. In this case, the addition of micaceous minerals to contaminated water bodies would serve as an effective in situ remediation technique by sequestering ^{137}Cs and reducing its bioavailability (Dabbagh *et al.*, 2008). The use of micaceous amendments (i.e., illites) to sequester contaminants, including ^{137}Cs , is not new. Most

previous methods, however, have physically mixed the amendment with the contaminated soil and subsequently tested the effectiveness using chemical adsorptive and/or desorptive tests.

The properties of 2:1 lattice clays (e.g., micas, vermiculites, illites, and smectites) lead to the almost completely irreversible adsorption of ^{137}Cs into the lattice structures. This process effectively immobilizes a large fraction of ^{137}Cs if sufficient time is allowed for equilibration. This concept led to the idea of adding illite to a ^{137}Cs -contaminated wetland to reduce concentrations in the biota (Singh, 2006). Replicated 3 m diameter limnocorrals were used to field test this method for different application rates of illite. Spreading of the illite on the water surface such that a depth of ~ 0.25 cm settled and covered the contaminated sediments resulted in a 25- to 30-fold reduction in ^{137}Cs concentrations in the water. Concomitant reductions in ^{137}Cs concentrations in plants were two- to fivefold and two- to threefold in fish (Strezov et al., 2009). This simple method reduced the biological availability of ^{137}Cs and preserved the integrity and function of the wetland at a cost that was 5–20 times less than that of sediment removal.

5.3.3 ADSORPTION

^{137}Cs sorption occurs primarily by ion exchange to the phyllosilicate fraction of soil or sediment. Experimentally measured ^{137}Cs -exchange isotherms often imply the presence of two or more sites with distinctive exchange energies (Brouwer et al., 1983; Cornell, 1993; Sawhney, 1972; Zachara et al., 2002). The high-energy site is believed to be associated with frayed-edge regions of weathered 1.0 nm micaceous minerals, and the low energy sites with the basal planes of expansible phyllosilicates including smectite and vermiculite. The presence of multiple sorption sites has been generally confirmed by NMR studies of ^{137}Cs -exchanged clay minerals (Kim et al., 1996; Weiss et al., 1990a, Weiss et al., 1990b). The short-term sorption/exchange of ^{137}Cs often exhibits reversible behavior (Brouwer et al., 1983; Zachara et al., 2002) and has been described by multisite, equilibrium exchange models (Brouwer et al., 1983; Cremers et al., 1988; Poinssot et al., 1999; Steefel et al., 2002; Zachara et al., 2002). The sorption and desorption of ^{137}Cs can exhibit kinetic behavior over longer contact periods (Comans et al., 1991; De-Preter, 1990; Di Toro, 1986; Evans et al., 1983). Slow diffusion of ^{137}Cs within edge-interlayer regions of micaceous minerals, accentuated by layer collapse around the poorly hydrated ^{137}Cs ion, is believed to cause this kinetic phenomenon (Eberl, 1980; Sawhney, 1967, 1969, 1972). Such diffusion may take days, months, years, or even decades for sorption/desorption to reach the equilibrium state depending on the structure of the mineral sorbents and the aqueous composition (Comans et al., 1991; De-Preter, 1990; Di Toro, 1986; Evans et al., 1983).

Edge and/or interlayer collapse on phyllosilicate sorbents slows ^{137}Cs desorption leading to exchange irreversibility or fixation (Comans et al.,

1991; De-Preter, 1990; Evans et al., 1983; Sawhney, 1972). Debate exists whether “fixed” ^{137}Cs is, or is not, exchangeable at slow rate (Comans et al., 1991; Cornell, 1993). The sorption of other exchangeable cations with low hydration energy, such as K^+ , Rb^+ , and NH_4^+ , may also collapse the edge-interlayer region, decreasing ^{137}Cs ion adsorption and desorption rate (Le Roux et al., 1970; Sawhney, 1967, 1972). In contrast, strongly hydrated cations (e.g., Na^+ and Ca^{2+}) may expand the edge-interlayer region—inducing an opposite effect.

The phenomenological aspects of ^{137}Cs desorption are understood primarily from investigations of single-phase 2:1 layer silicates. Comparable studies of ^{137}Cs contaminated soil and sediment are relatively rare. Long-term exposure allows ^{137}Cs diffusion into edge-interlayer regions of mica-ceous sorbents.

Limited study suggests that desorption rate and extent decreases with contact time (Evans et al., 1983; Smith and Comans, 1996), but controlling features and parameters are not understood. Understanding ^{137}Cs desorption from natural sediments subjected to long-term contamination is key to predicting future ^{137}Cs migration.

5.2.4 SOIL WASHING

Soil washing or the separation of soils by particle size may be an appropriate removal technique because ^{137}Cs is known to preferentially attach to small particles, especially clays (Adriano, 2004). This method, however, is generally only effective for soils that contain less than $\sim 20\%$ fine particles. Other sorting techniques can be used to physically separate soils that contain an activity level above a given threshold. For example, soil passes by radiation detectors on a conveyor belt, and when high activities are detected, that segment of soil is mechanically removed, whereas the low-activity soil continues on and is discarded as clean (Whicker et al., 2004).

6. FUTURE RESEARCH FOR ENVIRONMENTAL CONSERVATION

The 2011 Fukushima nuclear accident in Japan was the worst nuclear disaster following the 1986 Chernobyl accident. The FNPP accident, which was caused by the catastrophic earthquake and tsunami that occurred on March 11, 2011, led to large-scale emissions of radioactive ^{137}Cs and serious soil contamination around the FDNPP and in the neighboring prefectures. Therefore, if Fukushima-derived radionuclides were introduced into and above the planetary boundary layer, their dispersal on both regional and global scales would be inevitable. Indeed, the fission products released from the accidents have since spread not only across the entire northern hemisphere but also into the southern hemisphere (data available from the CTBTO Preparatory

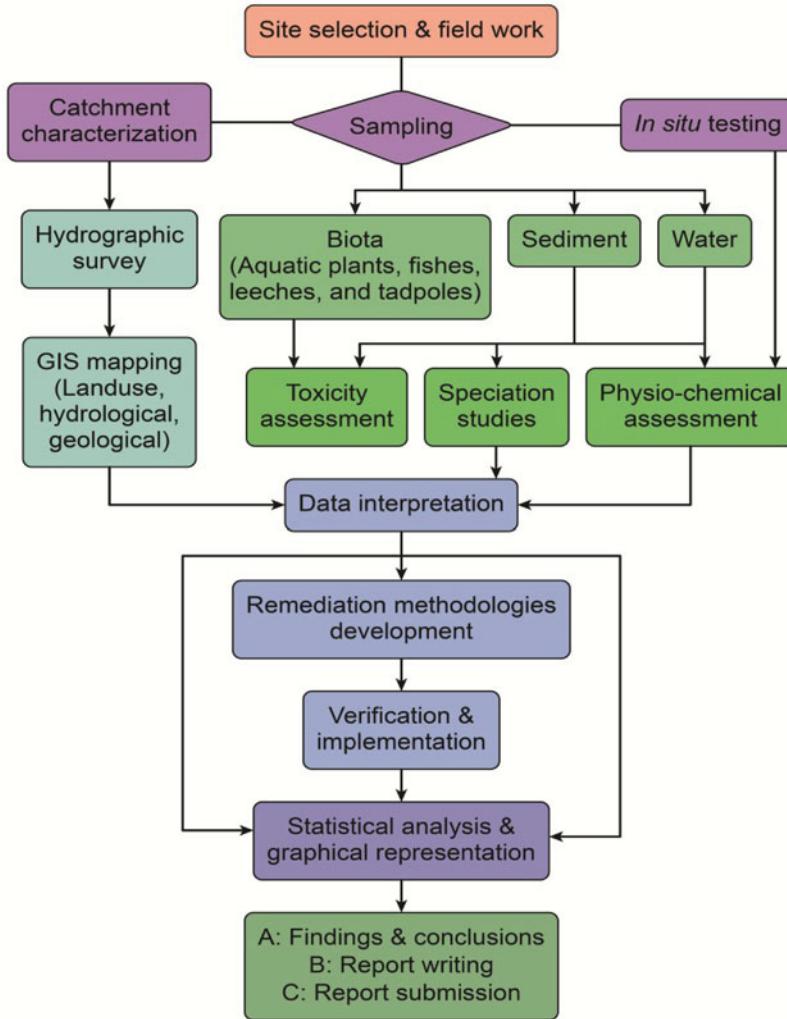


FIGURE 4. Flow sheet diagram for the future work plan.

Commission). After approximately two weeks, 15 fission products (nuclides) released from the Fukushima plant site had been detected around the globe.

The areas contaminated by radionuclides discharged from the Fukushima Daiichi nuclear power plant accident have been mapped in detail. However, the fate and transport of ^{137}Cs in the aquatic ecosystem, which are critical for their removal from the environment, have not yet been revealed. This review provides basic information on the fate and transport of ^{137}Cs in aquatic ecosystems and may serve as a starting foundation on which to devise future work plans. Figure 4 explains the flow sheet diagram for future research work on this topic.

Such research has a significant impact on and a demonstrable contribution to society and the economy. The following paragraph provides a brief introduction to the potential range of impacts that can be generated from the future research work in this field.

The future research will provide the scientific and technical information required by governmental sectors and related organizations for evaluation and future planning after the events of the tsunami in March 2011, which resulted in the release of radionuclides at the Fukushima Daiichi power station. This research represents a worldwide academic advancement that addresses global issues created by radionuclides. Little is known of the fate and transport of ^{137}Cs in the aquatic environment. Therefore, this research will be a major contribution to our understanding of the mechanism of radionuclide transport in the ecosystem. The development and utilization of new and innovative methodologies, equipment and cross-disciplinary approaches will provide major contributions toward the development of this academic discipline. Furthermore, the research will help to train highly skilled researchers for future development and growth.

This future work on this research will also contribute to evidence-based policy-making; influence public policies and legislation at the local, regional, national, and international levels; and enhance cultural enrichment, quality of life, health, and well-being. Additionally, this work will contribute to environmental sustainability, protection and impact reduction, and increased public awareness and understanding of science, economic, and social issues.

7. CONCLUSION

The evidence discussed in this review article indicates that ^{137}Cs may persist in a biologically available form for many years following its release into the environment. The high solubility of cesium and its major existence as ^{137}Cs in aqueous solution dictate a high degree of mobility and bioavailability. Moreover, ^{137}Cs displays chemical properties similar to other alkali metal cations with a similar ionic radius and charge, particularly the biologically essential K^+ . Thus, Cs^+ may be taken up via transport systems that catalyze the intracellular accumulation of K^+ under normal circumstances, and toxic effects of ^{137}Cs may result from the perturbation of K^+ -mediated processes. Despite these properties, the mobility of ^{137}Cs is strongly influenced by a number of environmental conditions and processes, and this appears to be particularly true in the case of aquatic ecosystems. One factor that is critical to determining cesium mobility in the environment is its tendency to become strongly complexed with inorganic components of sediments. This behavior contrasts markedly with its weak coordination with organic ligands. The complexation of ^{137}Cs with inorganics coincides with a dramatic reduction in its bioavailability. However, the binding of ^{137}Cs to suspended solids is not

necessarily irreversible, and this is particularly the case in aquatic ecosystems, where recycling of the bound ^{137}Cs may result from changes in the redox conditions of the water column, for example. The remediation of aquatic systems following contamination by ^{137}Cs is largely dependent on its target- and site-specific parameters. The effects of such remediation efforts, therefore, must be measured according to the specific environmental conditions. The remediation of contaminated ecosystem may involve physical, chemical, and biological procedures that each offer advantages and disadvantages. This review provides basic information on the fate and transport of ^{137}Cs in aquatic ecosystems and may serve as a key resource for devising future work plans in the wake of the FNPP disaster of 2011 and future unexpected environmental contamination by radiocesium.

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